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# Evidence for intrinsic magnetic scatterers in the topological semimetal $(Bi_2)_{s}(Bi_2Se_3)_{7}$

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# ARTICLE

# Evidence for intrinsic magnetic scatterers in the topological semimetal (Bi<sub>2</sub>)<sub>5</sub>(Bi<sub>2</sub>Se<sub>3</sub>)<sub>7</sub>

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## ABSTRACT

We report the synthesis and characterization of high-quality thin films of the topological semimetal (Bi<sub>2</sub>)<sub>5</sub>(Bi<sub>2</sub>Se<sub>3</sub>)<sub>7</sub>. Cryogenic magnetotransport experiments reveal strong metallic character and spin–orbit coupling in the films. By studying the temperature dependence of the electrical resistance of the topological semimetal, we observe a pronounced Kondo effect, which points toward the presence of magnetic scatterers. With the aid of density functional theory calculations, we identify Bi vacancies as intrinsic magnetic scatterers in this topological semimetal.

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# INTRODUCTION

Topological materials are exotic quantum materials with a nontrival band topology. They could be promising building blocks for future thin-film electronic devices with potential applications ranging from energy efficient spin-orbit torque memories,<sup>1</sup> to energy harvesting devices,<sup>2,3</sup> to neuromorphic computing devices,<sup>4</sup> or to a new generation of amplifier devices.<sup>5</sup> Recently, a new model system to study topological break-down and interlayer interactions has been discovered,  $(Bi_2)_n(Bi_2Se_3)_m$ , a natural hetero-structure of the model 3D topological insulator Bi<sub>2</sub>Se<sub>3</sub> and the 2D topological insulator Bi<sub>2</sub>. In bulk, it is a semimetal with Dirac-cone-like topological surface states that depend on its termination.<sup>6,7</sup> Furthermore, the compound could be very valuable for future functional electronics since it possesses topological surface states even in the bi-laver limit, enabling the fabrication of ultra-thin topological devices,<sup>8</sup> and it could be promising for energy harvesting applications thanks to its low thermal conductivity.9 Here, we experimentally reveal the presence of magnetic scatterers in thin films of  $(Bi_2)_5(Bi_2Se_3)_7$  by performing low-temperature magnetotransport experiments combined with indepth physical analysis. Supporting first-principle simulations allow us to identify Bi vacancies as magnetic scatterers in this topological semimetal.

High-quality (Bi<sub>2</sub>)<sub>5</sub>(Bi<sub>2</sub>Se<sub>3</sub>)<sub>7</sub> thin films were grown using a plasma-assisted molecular beam epitaxy (PA-MBE) technique on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>(0 0 0 1) c axis sapphire substrates (see the supplementary material). This hybrid approach has already been successfully used for the synthesis of transition metal dichalcogenide monolayers.<sup>10,11</sup> The exact composition of this hetero-structure has been determined by Rutherford backscattering spectrometry, Raman microscopy, and high-resolution transmission electron microscopy (see the supplementary material). After growth and in order to prevent surface degradation,<sup>12</sup> a thin (2 nm) capping layer of Al<sub>2</sub>O<sub>3</sub> or CaF<sub>2</sub> was deposited *in situ*. No influence of the type of the capping layer on the electronic properties of the thin films was found.

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**FIG. 1.** (a) Optical micrograph of a typical device. A current *I* is applied between the source and drain lead and the resulting voltages  $V_{xx}$  and  $V_{xy}$  are measured. Scale bar: 40  $\mu$ m. (b) Temperature dependent longitudinal resistivity  $\rho_{xx}$ . (c) Longitudinal and (d) transverse resistivity as a function of magnetic field measured at T = 2 K.

The as-grown films were patterned into Hall bar shape using standard optical lithography and dry etching/ion milling. An optical micrograph of a typical Hall bar device with dimensions  $L = 100 \,\mu\text{m}$  and  $W = 10 \,\mu\text{m}$  is shown in Fig. 1(a). The temperature and magnetic field dependent longitudinal ( $R_{xx}$ ) and transverse ( $R_{xy}$ ) resistances were obtained by applying a DC current of  $I = 1-10 \,\mu\text{A}$  to the source and drain leads of the device and by measuring the resulting DC voltage drops  $V_{xx}$  and  $V_{xy}$  [Fig. 1(a)].

We studied the magnetotransport behavior of ten devices fabricated using thin (Bi<sub>2</sub>)<sub>5</sub>(Bi<sub>2</sub>Se<sub>3</sub>)<sub>7</sub> films with a thickness of 12 nm from multiple MBE runs under identical growth conditions. Figure 1(b) shows the representative temperature dependent resistivity  $\rho_{rr}(T)$ (sample B).  $\rho_{xx}$  decreases with decreasing temperature, which can be attributed to a reduction of electron-phonon interaction and which is a characteristic of metallic samples.<sup>13</sup> At low temperatures (T < 20 K), a resistance minimum can be observed [see Fig. 2(a)]. We attribute this to Kondo correlations<sup>14</sup> present in the  $(Bi_2)_5$ (Bi<sub>2</sub>Se<sub>3</sub>)<sub>7</sub> films [see Fig. 2(a) and discussion below]. The metal-and 1(d) for such data on sample B]. The observed metallic behavior is in agreement with recent angle-resolved photo electron microscopy experiments, which found that natural heterostructures of Bi-bilayers and Bi2Se3 quintuple layers are bulk semi-metals that possess metallic, topological surface states.<sup>6,15</sup> The magnetoresistance is quadratic at high fields and shows a cusp due to weakantilocalization at low fields (see discussion in the supplementary material).

We further note a non-linearity in  $\rho_{xy}(B)$  measurements [Fig. 1(d)]. This could indicate transport via different electronic bands or spatially separated parallel conducting channels with different carrier mobilities and concentrations.<sup>16–18</sup>

Alternatively, as we will discuss below, this non-linearity in the Hall effect can indicate an anomalous Hall contribution to  $\rho_{xy}(B)$  induced by the presence of magnetic scatters inside our sample, similar to previous findings in magnetically doped Bi<sub>2</sub>Se<sub>3</sub>.<sup>19</sup>

In the following, we investigate the non-monotonic temperature dependence of the electrical resistance of  $(Bi_2)_5(Bi_2Se_3)_7$  films, which we attribute to the Kondo effect induced by local magnetic



**FIG. 2.** (a) Temperature-dependent longitudinal resistance of sample B (purple open circles) and a fit using Eq. (1) (green line). A field of B = 1 T was applied. (b) Normalized Kondo resistivity as a function of temperature (normalized by the Kondo temperature) for samples A–C (open circles) together with the universal functional predicted by NRG calculations (green line).

scatterers. Figure 2(a) shows the four-terminal resistance of sample B as a function of temperature measured at B = 1 T. This magnetic field was applied to suppress contributions from localization effects (WAL/WL)<sup>20</sup> and is well below the critical field  $B_c$  necessary to overcome the Kondo gap for a S = 1/2 system at  $T \ll T_K = 15$  K (see discussion below), where  $B_c = 0.5k_BT_K/(g\mu_B) \approx 6$  T.<sup>21</sup> We observe a logarithmic increase of  $R_{xx}$  below a temperature of 10 K followed by a saturation below 1 K. Such saturation disagrees with (anti-)localization effects where  $\partial R/\partial T \propto -\ln T$  for  $T \rightarrow 0$  and indicates the absence of a disorder-induced metal to insulator transition or the opening of a bandgap.<sup>20</sup>

In metallic systems containing magnetic impurities, the conduction electrons can couple anti-ferromagnetically to the local magnetic moments of the impurities. This enables a new spin-flip scattering process with an anomalous component of the resistance  $R_K(T/T_K)$ , which is approximately logarithmic in T when  $T \approx T_K$ . Below this characteristic Kondo temperature  $T_K$ , thermal fluctuations become weaker than the exchange energy and a so-called Kondo cloud is formed in which conduction electrons screen the local magnetic impurities. This leads to a saturation of the resistance. To this end, the temperature dependent resistance of the sample can be modeled as

$$R(T) = R_0 + qT^2 + pT^5 + R_{\rm K} \left(\frac{T}{T_{\rm K}}\right),\tag{1}$$

where  $R_0$  is the residual resistance due to sample disorder and the terms proportional to  $T^2$  and  $T^5$  describe contributions by electron–electron and electron–phonon interactions.<sup>22</sup> The contribution of the Kondo effect is described by the following empirical formula:

$$R_{\rm K}\left(\frac{T}{T_{\rm K}}\right) = R_{\rm K}(0) \left(\frac{T_{\rm K}^{\prime 2}}{T^2 + T_{\rm K}^{\prime 2}}\right)^{s},\tag{2}$$

where  $T'_{\rm K} = T_{\rm K}/(2^{1/s} - 1)^{1/2}$ . Assuming magnetic impurities with spin *S* = 1/2, the numerical factor becomes *s* = 0.225.

A fit to our experimental data using Eq. (1) is shown in Fig. 2(a) and yields  $R_0 = 2330 \ \Omega$ ,  $q = 2.4 \times 10^{-2} \ \Omega/\text{K}^2$ ,  $p = 1.6 \times 10^{-6} \ \Omega/\text{K}^5$ ,  $R_{\rm K}(0) = 30.8 \ \Omega$ , and  $T_{\rm K} = 16 \ \text{K}$ . These parameters can be used to re-scale the experimental data of various samples and compare their normalized Kondo resistivity  $R_{\rm K}(T)/R_{\rm K}(0)$  vs  $T/T_{\rm K}$  to the universal Kondo behavior from numerical renormalization group calculations (NRG).<sup>23</sup> Such scaling is shown Fig. 2(b). We observe that all experimental curves follow an universal functional and that this functional can be well described by the NRG calculations for the Kondo effect.

In order to find the origin of magnetic scatterers in our sample, we performed first-principles simulations, based on density functional theory (DFT), of  $Bi_2/(Bi_2Se_3)_n$  stacks, including different intrinsic point defects. The atomic configuration and energy band structure of Bi<sub>2</sub>/Bi<sub>2</sub>Se<sub>3</sub> are shown in Figs. 3(a) and 3(b), respectively. The system is predicted to be semi-metallic, in agreement with other DFT calculations reported in the literature.<sup>6,15</sup> Various intrinsic point defects in 2D Bi2/Bi2Se3 stacks were first investigated using  $(3 \times 3)$  supercells. We considered Bi  $(V_{Bi})$  and Se  $(V_{Se})$  vacancies and Bi (Bise) and Se (SeBi) antisites, present at different locations in the Bi<sub>2</sub>/Bi<sub>2</sub>Se<sub>3</sub> stack, as shown in Fig. S1 (see the supplementary material). All these defects have relatively low formation energies, lying typically between 0.5 and 2 eV (see Fig. S2), except for  $Se_{Bi}$ , which has a negative formation energy, with this defect being spontaneously formed in the 2D material. Surprisingly, none of these defects have a net magnetic moment; this can be attributed to the charge transfer occurring between the Bi2 and Bi2Se3 layers. Indeed, the Bi\_2 layer tends to give electrons (about  $7.8\times 10^{13} e/cm^2)$  to the Bi<sub>2</sub>Se<sub>3</sub> layer,<sup>24</sup> which leads to an empty (defect in Bi<sub>2</sub>) or fully occupied (defect in Bi2Se3) defect level, the defect then being nonmagnetic. As shown in the supplementary material (see Fig. S3), when the distance between the Bi2 and Bi2Se3 layers is artificially increased, which results in a reduced charge transfer between these layers, the magnetic moment of some defects "reappears." We next considered defects in a Bi<sub>2</sub>/(Bi<sub>2</sub>Se<sub>3</sub>)<sub>2</sub> stack. The calculated charge



FIG. 3. (a) Top and side views of the atomic structure of  $Bi_2/Bi_2Se_3$ . The green and purple spheres correspond to the Se and Bi atoms, respectively. (b) Energy band structure of  $Bi_2/Bi_2Se_3$ . The reference (zero) energy level corresponds to the Fermi level.



**FIG. 4.** (a) Atomic structure of a Bi<sub>2</sub>/(Bi<sub>2</sub>Se<sub>3</sub>)<sub>2</sub> stack with a Bi vacancy in the bottom Bi<sub>2</sub>Se<sub>3</sub> layer. The band-decomposed charge densities in the energy range between -0.02 and 0.03 eV are also shown in yellow. (b) Projected band structures of the system for spin-up and spin-down electrons. The red lines correspond to the contributions from the 4*p*-Se orbitals around the Bi vacancy.

transfer to the bottom Bi2Se3 layer is reduced by about an order of magnitude, as compared to the top Bi<sub>2</sub>Se<sub>3</sub> layer. We studied the same defects as discussed above, present at different possible sites in the bottom Bi2Se3 layer, and identified only one defect with a net magnetic moment of about 0.55  $\mu_{\rm B}$ , namely, a Bi vacancy. The atomic configuration of the corresponding defective structure is shown in Fig. 4(a). The formation energy of this Bi vacancy in the Bi<sub>2</sub>/(Bi<sub>2</sub>Se<sub>3</sub>)<sub>2</sub> structure lies between about 0.5 and 1 eV, in the Se-rich and Bi-rich limit, respectively. As shown in Fig. 4(b), localized states, corresponding to the 4p-Se orbitals neighboring the Bi vacancy site, are clearly observed in the electronic density of states within an energy range between 0 and 0.5 eV from the Fermi level  $E_{\rm F}$ . The spin-polarized energy band structure also indicates the presence of localized spin-down states near  $E_{\rm F}$ , associated with the 4p-Se orbitals. We thus tentatively assign the magnetic defects responsible for the Kondo effect observed in our samples to these Bi vacancies.

#### SUMMARY

To summarize, we present a hybrid epitaxy approach, PA-MBE, suitable to grow high-quality thin films of the topological semimetal  $(Bi_2)_5(Bi_2Se_3)_7$ . Depth cryogenic magnetotransport experiments combined with first-principle simulations allowed us to reveal that Bi vacancies could act as intrinsic magnetic scatterers in this material. These scatterers explain the observed pronounced Kondo effect in the temperature dependent longitudinal resistance of the devices. Therefore, our work highlights how intrinsic impurities can strongly impact topological properties, an important finding when considering topological materials for future device integration.

## SUPPLEMENTARY MATERIAL

The supplementary material contains experimental details about crystal growth and characterization (Raman, TEM, RHEED, XRD, RBS), atomic force microscopy, details about weakantilocalization and computational methods.

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# AUTHOR DECLARATIONS

#### **Conflict of Interest**

The authors have no conflicts to disclose.

#### Author Contributions

Pascal Gehring: Conceptualization (lead); Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (lead); Writing – original draft (lead). Clement Merckling: Formal analysis (equal); Investigation (equal); Methodology (equal); Writing – review & editing (equal). Ruishen Meng: Formal analysis (equal); Methodology (equal); Writing – review & editing (equal). Valentin Fonck: Formal analysis (equal); Validation (equal); Writing – review & editing (equal). Bart Raes: Methodology (equal); Writing – review & editing (equal). Michel Houssa: Supervision (equal); Writing – review & editing (equal). Joris Van de Vondel: Supervision (equal); Writing – review & editing (equal). Stefan De Gendt: Supervision (equal); Writing – review & editing (equal).

#### DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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